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(54) **Transparent and slippery biaxially stretched polyester film**

(57) A biaxially stretched polyester film having excellent transparency and slipperiness, containing 0.005 to 1% by weight of an organic lubricant and having the following properties after biaxial stretching and heat-setting:

$$\Delta H \leq 0.3\% \quad (1)$$

$$H_0^{100} \leq 0.9\% \quad (2)$$

$$\mu s \leq 1.2 \quad (3)$$

wherein ΔH is the surface haze of the biaxially stretched film, H_0^{100} is the internal haze of the film as calculated based on the 100 μm film thickness, and μs is the coefficient of static friction of the film. It is used as a based film for recording materials such as microfilms, printing plates, X-ray photographs, and for transparent electrode substrates.

GB 2 188 586 A

SPECIFICATION

Transparent and slippery biaxially stretched polyester film

- 5 The present invention relates to a biaxially stretched polyester film having excellent physical properties, especially slip properties and optical properties, and suited for use as a base film for recording materials such as photographs, printing plates, etc. and transparent electrode substrates and the like. More particularly, the present invention relates to a base film having excellent slip properties and transparency as well as good adhesive property of the printed layer or deposited film formed on the base film and high durability thereto in use for recording material such as printing plates, X-ray photographs, microfilms, electrophotographs, diazo copies, etc., and transparent electrode substrates, etc., for which a film having, inter alia, excellent slip properties and transparency is required. 5
- 10 Polyester film is provided with almost all of the properties required for a base film for photographs, printing plates, transparent electrode substrates, etc.: 10
- 15 (1) to be transparent and cloudless, 15
 (2) to be resistant to tensile force, tear force and twist force,
 (3) to be dimensionally stable and not to be curled,
 (4) to be stable against emulsion, water and alkaline solutions,
 (5) to have a high thermal deformation temperature,
 20 (6) to be free of alien matter on the surface and in the inside of the film, etc. 20
- In recent years, however, demand has grown for a higher-quality base film for recording materials such as photographs, printing plates, etc. and transparent electrode substrates, etc., and it has been required to satisfy particularly the antinomic requirements of transparency and slipperiness to a higher degree.
- As means for improving the slip properties of polyester film, it has been tried to form an unevenness on the film surface with fine particles by, for instance, a so-called particle separation method in which the inactive fine particles formed from the catalyst residue are separated on the film surface or a particle addition method in which the fine particles of an inorganic compound are blended in the course of polymerization or melt extrusion. These conventional methods, however, had the problem that, as far as the conventional film-forming and stretching conditions are employed, when an unevenness is formed on the film surface to increase the surface roughness, the surface haze of the film rises and the voids produced at the interface between the fine particles and the polymer are increased. The internal haze is also raised, resulting in a reduced transparency of the film as a whole. 25
- 30 The present inventors previously found and disclosed in Japanese Patent Application No. 138339/85 that a film improved in transparency and slip properties could be obtained without affecting the thickness unevenness and flatness of the film by controlling the degree of surface orientation (ΔP) of the film, the average refractive index (\bar{n}) of the film and the x-ray defraction strength ratio of the plane (110) to the plane (100). In this proposal, however, the particles contained in the film were still a main factor of slipperiness, and it was very difficult to greatly reduce the content of the particles and drastically improve the transparency while maintaining the desired level of slipperiness. 35
- 40 Lately, the desire for the transparency of base film for recording material such as photographs, printing plates, etc. and transparent electrode substrates has become more intense, and the desire has reached such a point that no solution is possible unless the content of the particles is reduced greatly from the conventional level. The conventional films showing a very low content of particles and a high degree of transparency, and as a countermeasure, both sides of the film have been subjected to a mechanical working to produce unevenness thereon at the time of shipment so as to prevent the film from suffering scratches when it was wound up. However, irrespective of such processing, the film would suffer from many scratches in the film-forming process, and further the addition of the step of giving unevenness to both side of the film led to a rise in the manufacturing cost of the film. Thus, a method for producing a film of greatly improved transparency, while maintaining the good slip properties, by greatly reducing the particle content from the conventional level has been strongly desired. 45
- 50 In view of the above-mentioned problems, as a result of the present inventors' studies and experiments, it has been found that a film strikingly improved in transparency by greatly reducing the particle content could still maintain high slipperiness and excellent wind-up properties only when the film has from 0.005 to 1.0% by weight of an organic lubricant contained in the polyester and also meets the following relations (1) to (3) after biaxially stretching and heat-setting: 55

$$\begin{aligned} \Delta H &\leq 0.3\% & (1) \\ H_0^{100} &\leq 0.9\% & (2) \\ \mu_s &\leq 1.2 & (3) \end{aligned}$$

- 60 wherein ΔH is the surface haze of the biaxially stretched film H_0^{100} is the internal haze of the film as calculated based on the 100 μm thickness of the biaxially stretched film, and μ_s is the coefficient of static friction of the biaxially stretched film. The present invention was attained on the basis of this finding. 60

- In the first aspect of the present invention, there is provided a biaxially stretched polyester film having excellent transparency and slipperiness, containing from 0.005 to 1% by weight of an organic lubricant and 65

having the following properties after biaxially stretching and heat-setting:

- 5 $\Delta H \leq 0.3\%$ (1)
 $H_0^{100} \leq 0.9\%$ (2)
 $\mu s \leq 1.2$ (3) 5

wherein ΔH is surface haze of the biaxially stretched film, H_0^{100} is internal haze of the film as calculated based on the 100 μm film thickness, and μs is coefficient of static friction of the film.

- 10 In a second aspect of the present invention, there is provided a recording material comprising a biaxially stretched film containing from 0.005 to 1% weight of an organic lubricant and having the following properties: 10

- 15 $\Delta H \leq 0.3\%$
 $H_0^{100} \leq 0.9\%$
 $\mu s \leq 1.2$ 15

(wherein ΔH is surface haze of the biaxially stretched film, H_0^{100} is internal haze of the film as calculated based on the 100 μm film thickness, and μs is coefficient of static friction of the biaxially stretched film), and at least one of a photosensitive layer, an emulsion layer, a transparent conductor layer having a photoconductor layer or a photosensitive layer laminated on the polyester film.

- 20 In a third aspect of the present invention, there is provided a transparent electrode comprising a biaxially stretched film containing from 0.005 to 1% by weight of an organic lubricant and having the following properties: 20

- 25 $\Delta H \leq 0.3\%$
 $H_0^{100} \leq 0.9\%$
 $\mu s \leq 1.2$ 25

- 30 (wherein ΔH is surface haze of the biaxially stretched film, H_0^{100} is internal haze of the film as calculated based on the 100 μm film thickness, and μs is coefficient of static friction of the biaxially stretched film), and a transparent conductor layer laminated on the film. 30

The heart of the present invention is characterized in that a biaxially stretched polyester film showing excellent transparency and slip properties contains 0.005 to 1% by weight of an organic lubricant and meets the following relations (1) to (3) after biaxially stretching and heatsetting:

- 35 $\Delta H \leq 0.3\%$ (1) 35
 $H_0^{100} \leq 0.9\%$ (2)
 $\mu s \leq 1.2$ (3)

- 40 wherein ΔH is the surface haze of the biaxially stretched film, H_0^{100} is the internal haze of the film as calculated based on the 100 μm thickness of the biaxially stretched film, and μs is the coefficient of static friction of the biaxially stretched film. 40

- 45 The term "polyester" used in the present invention refers to the polymers obtainable by polycondensing aromatic dicarboxylic acids such as terephthalic acid, isophthalic acid, and naphthalene-2,6-dicarboxylic acid, or esters thereof and glycols such as ethylene glycol, diethylene glycol, tetramethylene glycol, and neopentyl glycol. 45

- Such polyesters can be obtained by various methods such as mentioned below:
 an aromatic dicarboxylic acid and a glycol are directly polycondensed;
 a dialkyl ester of an aromatic dicarboxylic acid and a glycol are subjected to an ester exchange reaction and then polycondensed; and
 50 a diglycol ester of an aromatic dicarboxylic acid is polycondensed. 50

- Typical examples of such polymers are polyethylene terephthalate and polyethylene-2,6-naphthalene dicarboxylate. Such polymers may be non-copolymerized homopolymers or may be copolymerized polyesters in which not more than 15% by mole of the dicarboxylic acid component is constituted by a non-aromatic dicarboxylic acid and/or not more than 15% by mole of the diol component is constituted by a diol other than aliphatic glycols. 55

It is also possible to use polyblends of the above-mentioned polyesters and other polymers such as polyamides, polyolefins and other types of polyesters (such as polycarbonates).

It is also advisable to include a polyalkylene glycol for the purpose of improving printability of the film or adhesive property to the deposit thereon.

- 60 As polyalkylene glycols, polyethylene glycol, polytetramethylene glycol, propylene glycol may be exemplified. Various methods are available for introducing such polyalkylene glycol to the film. For instance, it may be introduced in the course of ester exchange or polymerization, or a polymer produced by copolymerizing a polyalkylene glycol may be blended in the film composition or may be incorporated in the composition at the time of drying or extrusion. As for the molecular weight of such polyalkylene glycol, it is preferably not greater than 10,000, more preferably not greater than 8,000, in order that the transparency of the produced film shall 65

not be affected by the addition of such polyalkylene glycol. The content of polyalkylene glycol in the film is preferably not greater than 1%, more preferably not greater than 0.5%.

The polyester may further contain at least one additive such as a stabiliser, coloring agent, antioxidant, defoaming agent, etc., as desired. It is also possible to add fine particles and/or inactive inorganic particles to further increase the slipperiness of the polyester film.

The fine particles can be inactive external particles such as kaolin, clay, calcium carbonate, silicon oxide, calcium terephthalate, aluminum oxide, calcium phosphate, titanium oxide, titanium oxide, or other salts or oxides containing the elements selected from Groups I, II, III, IV, etc., of the Periodic Table, or internal particles, formed into the polymer in the course of production of polyester by a high-melting point organic compound insoluble at the time of melting and film-forming of polyester resin, or a metal compound catalyst used for the synthesis of crosslinked polymers or polyesters, such as alkali metal compounds or alkali earth metal compounds.

The amount of the fine particles contained in the film should be from 0.0001 to 0.5% by weight, preferably from 0.001 to 0.05 by weight. Also, these particles should have an average particle size in the range of from 0.01 to 3.5 μm , preferably from 0.5 to 2.0 μm .

In case of using the fine particles, it is preferable that they are of a bimodal system consisting of large particles and small particles.

In the present invention, it is essential that the surface haze (ΔH) of the film after biaxially stretching and heat-setting is not more than 0.3%, preferably from 0.05 to 0.25%, and the internal haze (H_0^{100}) of the film as calculated based on the 100 μm thickness of the film is not more than 0.9%, preferably from 0.2 to 0.8%. If the surface haze (ΔH) is more than 0.3%, the irregular reflection at the surface of the film is increased to spoil the transparent appearance of the film. If the internal haze (H_0^{100}) becomes more than 0.9%, light absorption in the inside of the film augments excessively, which also spoils the transparent appearance of the film. It is preferable that the total haze (H_1^{100}) of the film as calculated based on the 100 μm film thickness is not more than 1.2%, preferably not more than 1.0%, as the transparency of the whole film is enhanced when H_1^{100} is in this range.

The coefficient of inter-film static friction (μ_s) can serve as a measure for knowing the easiness of film wind-up at the completion of film formation or at the time of slitting. In the present invention, it is essential that μ_s of the film after biaxial stretching and heat-setting is not greater than 1.2, preferably not greater than 1.0. If μ_s is greater than 1.2, granular marks due to blocking tend to be produced on the film roll surface when the film is wound up after completion of film formation or at the time of slitting, making the film unable to stand practical use. It is preferable in terms of film wind-up properties that the coefficient of kinetic friction (μ_d) of the film is not greater than 1.2 and smaller than μ_s .

For satisfying the above-mentioned both requirements for high-degree transparency and slipperiness, it is insufficient to add the fine particles and/or inactive inorganic particles to the polyester, but it is necessary also to add an organic lubricant to the polyester. As such organic lubricant, it is preferred to use a bisamide compound represented by the following general formula (4) or (5) as this compound contributes greatly to the slipperiness of the film and also has little effect on the haze of the film.



In the above formulae, R_1 represents an alkylene group having 1 to 12 carbon atoms, such as methylene, ethylene, trimethylene, tetramethylene, hexamethylene, octamethylene, decamethylene or dodecamethylene. Alkylene groups having 1 to 6 carbon atoms are preferred.

R_2 and R_3 each independently represents an alkyl group having 8 to 30 carbon atoms, such as octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, nonadecyl, icosyl, heneicosyl, docosyl, heptacosyl or triacontyl. Alkyl groups having 12 to 30 carbon atoms are preferred.

Each of the R_1 to R_3 may have a branched chain.

R_4 represents hydrogen or a methyl group.

The bisamide compounds represented by the general formula (4) can be obtained by, for instance, reacting alkylenediamines such as ethylenediamine, propylenediamine, butylenediamine, hexamethylenediamine, octamethylenediamine, decamethylenediamine, or dodecamethylenediamine with aliphatic acids such as stearic acid, capric acid, decanoic acid, lauric acid, myristic acid, palmitic acid, arachidic acid, behenic acid, oleic acid, elaidic acid, montanic acid, or carnaubic acid. Typical examples of such bisamide compounds are

N,N'-methylenebisstearyl acid amide, N,N'-thylenebisstearyl acid amide, N,N'-butylenebisstearyl acid amide, N,N'-butylenebismyristic acid amide, and N,N'-hexamethylenebisbenzoic acid amide.

In the formula (5), R_5 represents an alkylene group having 1 to 12 carbon atoms or a divalent hydrocarbon residue containing an aromatic ring or aliphatic ring. Examples thereof are allylene groups such as phenylene group and naphthylene group; allylenedialkylene groups in which two hydrogen atoms of an aromatic hydrocarbon are substituted with two alkylene groups, such as xylylene group; diallylenetrialkylene groups such as diphenylenetrialkylene group; arylsubstituted alkylene groups; and cycloalkylene groups such as cyclohexylene group and cyclopentylene group. Cycloalkylene groups having 6 to 18 carbon atoms are preferred. These groups may have a substituent or substituents.

- 10 The bisamide compounds represented by the general formula (5) can be obtained by, for instance, reacting alkylmonoamines such as octylamine, decylamine, pentadecylamine, heptadecylamine, octadecylamine, and dodecylamine with carboxylic acids such as cyclohexanedicarboxylic acid, succinic acid, adipic acid, sebacic acid, terephthalic acid, isophthalic acid, phenylethyldicarboxylic acid, phenylethanedicarboxylic acid, naphthalenedicarboxylic acid, and biphenyldicarboxylic acid. Typical examples of such bisamide compounds are
- 15 N,N'-diheptadecyladipic acid amide, N,N'-diheptadecylsebacic acid amide, N,N'-diheptadecylterephthalic acid amide and N,N'-diheneicosylterephthalic acid amide.

These bisamide compounds, both those represented by the formula (4) and those represented by the formula (5), may be used either singly or as a mixture.

- The content of such an organic lubricant in the film is from 0.005 to 1% by weight, preferably from 0.005 to 0.5% by weight, more preferably from 0.01 to 0.5% by weight. If it is less than 0.005% by weight, the effect of the compound for improving the slip properties of the film is unsatisfactory. If the content exceeds 1% by weight, the deposition of the organic lubricant on the film surface becomes excessive, resulting in a worsened film haze or a reduction of adhesion and durability of the printed layer or deposited film on the polyester film.

- As for the time of adding the bisamide in the production of the transparent slippery polyester film of the present invention, it is preferred to add the bisamide compound at a stage after the preparation of polyester resin and prior to extrusion molding. For instance, the compound may be directly dispersed and mixed in the polyester resin prior to extrusion molding, or a high-concentration masterbatch resin may be prepared earlier and this masterbatch is mixed with a polyester homopolymer to a desired concentration, and then extrusion molding is carried out to form a film. It is possible to employ methods other than the incorporation method,
- 30 for example, a method in which the compound is added after completion of the polycondensation reaction.

Also, as a result of many studies on the conditions under which the organic lubricant can best contribute to the slipperiness of the film after biaxially stretching and heat-setting, when the lubricant content in the film is fixed, it was found that the average refractive index (\bar{n}) of the film is closely associated with the slipperiness of the produced film.

- 35 It is preferable that in order to maximise the contribution of the organic lubricant to the slipperiness of the film, the average refractive index (\bar{n}) of the film after biaxially stretching and heat-setting is in the range of 1.640 to 1.670, preferably 1.650 to 1.670. If \bar{n} is less than 1.640, the organic lubricant tends to be unable to make a sufficient contribution to the slipperiness of the film. If \bar{n} exceeds 1.670, although desired slipperiness of the film can be obtained, there is a tendency for the crystallization of the film to advance thereby adversely effecting the transparency and strength of the film. It is difficult to give a correct account of the interrelationship between \bar{n} and slipperiness, but the following assumption appears credible.
- 40

It is generally considered that an organic lubricant such as mentioned above is concentrated in the amorphous phase of polyester film, so that it is supposed that by increasing \bar{n} , that is, by enhancing the crystallization degree of the film, the organic lubricant is allowed to deposit effectively on the film surface.

- 45 Further, quite surprisingly, the fact is noted that in the case of a base film which has been improved in slipperiness by merely containing a large amount of the organic lubricant, without \bar{n} of the film being defined in the above-mentioned range, the printed film formed on the base film tends to be reduced in adhesion and durability. Whereas in the case of a film which has attained its desired slipperiness with the minimum necessary content of the organic lubricant and with \bar{n} being defined within the above-mentioned range, there occurs almost no reduction of adhesion and durability of the printed layer or deposited film. It seems that this fact owes to the very effective deposition, in a finely dispersed state, of the organic lubricant on the film surface in the latter film. It is thus considered that the method in which the organic lubricant is deposited on the film surface while increasing the crystallization degree of the film makes it far easier to control the deposit and dispersed state of the organic lubricant on the film surface and is also capable of producing a stronger
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- 55 bond between the film and the organic lubricant than possible with the method in which the organic lubricant is merely contained in a large amount.

In order to obtain preferable thickness unevenness and flatness while maintaining high slipperiness of the film, the surface orientation (ΔP) of the film after biaxially stretching and heat-setting is in the range of 0.155 to 0.175, preferably 0.155 to 0.170. If ΔP is less than 0.155, it tends to be impossible to maintain the thickness unevenness and flatness of the film. Also, the haze of the film tends to increase, that is, the transparency is worsened, with decrease of ΔP . If ΔP exceeds 0.175, the slipperiness of the film tends to be reduced.

- As described above, a very excellent biaxially stretched polyester film having both high-degree transparency and slipperiness and advantageously usable as film base for recording materials such as printing plates, X-ray photographic plates, microfilms, electrophotographic plates, diazo photographic plates, etc., and transparent electrode substrates, etc., can be obtained by containing an organic lubricant in an amount of
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0.005 to 1% by weight in the polyester and adjusting ΔH , H_0^{100} and μ_s of the film after biaxially stretching and heat-setting so that they fall in the specified ranges shown above.

The process of forming the polyester film according to the present invention will be described in further detail below.

- 5 The polyester chips are mixed with fine particles of kaolin, silica or the like and an organic lubricant, and, if necessary, with such additives as stabilizer, coloring agent, defoaming agent, polyalkylene glycol, etc., are dried by the usual means, then extruded through an extruder and cooled and solidified on a rotating cooling body to form a non-stretched polyester sheet. For the cooling step, it is preferred to use the ordinary electrostatic cooling method. The thus obtained film is first stretched in the first axial direction, usually in the machine direction by from 3.0 to 5.0 time the original length, so that the birefringence index thereof will exceed 0.070. The stretching temperature is preferably between 75 and 130°C. As the rolls used for such stretching, any suitable types of rolls such as hard chrome-plated rolls, ceramic rolls, elastomer rolls such as Teflon rolls, etc., can be properly used. Then the monoaxially oriented film, either after cooling below the glass transition temperature or without cooling and then pre-heating to a temperature of 90–150°C, is further stretched in the second axial direction (the transverse direction) by 3.2 to 5.0 times the original width at substantially same temperature to obtain a biaxially oriented film. If the stretching ratio in the second axial direction is less than 3.2 times, the thickness unevenness in the second axial direction will become too large. This biaxially oriented film is then heat-set at 200–250°C for a period of 1 second to 10 minutes. Preferably, the film is relaxed 1 to 15% in the widthwise direction at 200°C or above in the heat-setting zone. Most preferably, the film is relaxed in the widthwise direction in the maximum heat-set temperature zone.

It is also a preferred form of practice to employ a so-called double heat-setting method in which the film, after heat-setting at not less than 200°C, is cooled to not more than 120°C and then again heat-set at not less than 200°C for the purpose of further enhancing the transparency and slipperiness of the film.

- The biaxially stretched film according to the present invention is usable as base film for recording materials such as printing plates, X-ray photographic plates, microfilms, electrophotographic plates and diazo photographic plates, and transparent electrode plates. The structures of these articles incorporating the polyester film of the present invention are exemplified below.

- (1) A printing plate comprises a transparent and slippery biaxially stretched polyester film containing 0.005 to 1% by weight of an organic lubricant and having a surface haze of not more than 0.3%, an internal haze of not more than 0.9% as calculated based on the 100 μm film thickness and a coefficient of static friction of not more than 1.2, and at least a known type of photosensitive layer laminated on to the polyester film, if necessary with a known adhesive layer or a known pigmented adhesive layer disposed therebetween.

- (2) An X-ray photographic plate comprises a transparent and slippery biaxially stretched polyester film containing 0.005 to 1% by weight of an organic lubricant and having a surface haze of not more than 0.3%, an internal haze of not more than 0.9% as calculated based on the 100 μm film thickness and a coefficient of static friction of not more than 1.2, and at least a known type of emulsion layer laminated on the polyester film, if necessary with a known adhesive layer disposed therebetween.

- (3) A microfilm or electrophotographic plate comprises a transparent and slippery biaxially stretched polyester film containing 0.005 to 1% by weight of an organic lubricant and having a surface haze of not more than 0.3%, an internal haze of not more than 0.9% as calculated based on the 100 μm film thickness and a coefficient of static friction of not more than 1.2, a known transparent conductor layer laminated on the biaxially stretched polyester film, and a known photconductor layer laminated on the transparent conductor layer.

- (4) A diazo photographic plate comprises a transparent and slippery biaxially stretched polyester film containing 0.005 to 1% by weight of an organic lubricant and having a surface haze of not more than 0.3%, an internal haze of not more than 0.9% as calculated based on the 100 μm film thickness and a coefficient of static friction of not more than 1.2, and a known photosensitive layer containing a diazo compound, a coupler and an assistant (or assistants), the photosensitive layer being laminated on the polyester film, if necessary, after its surface has been subjected to corona discharge or other typical chemical treatment.

- (5) A transparent electrode comprises a transparent and slippery biaxially stretched polyester film containing 0.005 to 1% by weight of an organic lubricant and having a surface haze of not more than 0.3%, an internal haze of not more than 0.9% as calculated based on the 100 μm film thickness and a coefficient of static friction of not more than 1.2, and a known type of transparent conductor layer laminated on the polyester film.

- As described above in detail, in accordance with the present invention, there is provided a film having the transparency and slipperiness required in use for recording materials such as printing plates, X-ray photographic plates, microfilms, electrophotographic plates, diazo photographic plates, etc., transparent electrode substrates, etc., and also having good adhesive properties to the printed layer or the film deposited thereon to provide an excellent durability thereto. The thickness of the film of the present invention is preferably from 3 to 500 μm , preferably from 20 to 200 μm .

- Thus, the film according to the present invention is a polyester film having very excellent transparency and slipperiness and useful as a base film for photographs, printing plates, transparent electrode substrates, etc.

The present invention will hereinafter be described more particularly in accordance with the examples thereof, which examples are, however, given for the purpose of illustration and are not to be construed as limiting the scope of the present invention.

1. Film haze

The surface haze and internal haze of the film were determined according to JIS-K 6714 by using an integrating sphere type turbidimeter NDH-20D mfd. by Nippon Denshoku Kogyo KK.

The surface haze (ΔH) and the internal haze (H_0^{100}) and haze (H_1^{100}) as calculated based on the 100 μm thickness of the film are given from the following formulae:

$$\Delta H = H_1 - H_0$$

$$H_0^{100} = H_0 \times \frac{100}{d}$$

$$H_1^{100} = H_0^{100} + \Delta H$$

wherein d is the film thickness (μm), H_1 is the measured value of film haze, and H_0 is the measured value of the film haze after liquid paraffin has been applied to the film surface, as internal haze.

15 2. Average refractive index and degree of surface orientation

The refractive index of the film was measured by using an Abbe's refractometer mfd. by ATAGO Co., Ltd. A sodium lamp was used as light source.

The maximum refractive index (n_γ) in the film area, the refractive index (n_β) in the direction orthogonal thereto and the refractive index (n_α) in the thickness direction were measured, and the average refractive index and the degree of surface orientation were calculated from the following equations:

$$\text{Average refractive index} = \frac{n_\gamma + n_\beta + n_\alpha}{3}$$

$$\text{Degree of surface orientation} = \frac{n_\gamma + n_\beta}{2} - n_\alpha$$

3. Birefringence index (film stretched in machine direction)

The retardation was measured by a polarizing microscope mfd. by Carl Zeiss Corp., and the birefringence index (Δn) was determined from the following equation:

$$\Delta n = R/d$$

wherein R is retardation, and d is film thickness (μm).

35 4. Slip properties

Two films, placed one upon the other, were pailed on a smooth glass plate. A rubber plate was placed atop the film and further a weight was placed thereon so that the contact pressure between the two films would be 2 g/cm². The films were moved sliding against each other at a rate of 20 mm/min and the frictional force was measured. The coefficient of friction at the point where the films were slid 5 mm relative to each other was counted as coefficient of kinetic friction.

5. Intrinsic viscosity (η)

1 g of polymer was dissolved in 100 ml of a 50/50 (by weight) phenol/tetrachloroethane mixed solvent, and the viscosity of the solution was measured at 30°C.

6. Durability of metallized film

An approximately 1,500 Å thick indium oxide film was formed on a polyester film by vacuum evaporation, and two sheets of the laminate film were placed one upon the other while spacing apart from each other by a spacer to form a switch. The key was struck at 3 Hz by using a 200 gram-load rod having 7R rubber at the end under the conditions of electrode area: 2 cm × 2 cm, interval: 100 μm , and circuit current: 1 mA. The number of times the disturbance of the signal wave form occurred was counted, and the evaluation was made on the following criterion:

- O: More than 1,000,000 times. No problem at all for practical use.
 Δ: 300,000 to 1,000,000 times. Some problems involved for practical use.
 X: Less than 300,000 times. Cannot stand use.

EXAMPLES 1-2 & COMPARATIVE EXAMPLES 1-3

Preparation of polyester chips:

60 100 parts by weight of dimethyl terephthalate, 70 parts by weight of ethylene glycol and 0.07 parts by weight of calcium acetate monohydrate were supplied into a reactor and heated to carry out an ester exchange reaction while distilling off methanol. Heating was continued till it reached 230°C, about 4.5 hours after start of the reaction, at which point the ester exchange reaction was substantially completed.

Then, 0.04 part by weight of phosphoric acid and 0.035 part by weight of antimony trioxide were added, and the mixture was polymerized by an ordinary method. That is, the reaction temperature was raised gradually

so that it finally reached 280°C while the pressure was reduced gradually so that it finally reached 0.5 mmHg. The reaction was terminated after 4 hours, and the reaction product was made into chips in the usual way to obtain polyester (A).

In the meantime, the same process as the preparation of polyester (A) was carried out except that 0.10 part by weight of amorphous silica of fine particles having an average diameter of 1.3 μ was added after the end of the ester exchange reaction to obtain polyester (B). 5

In a separate operation, 98 parts by weight of polyester (A) was dried in vacuo at 180°C for 6 hours, and after cooling the same, 2 parts by weight of fine powder of N,N'-hexamethylenebisstearic acid amide was added and incorporated at 280°C by using an extruder to obtain polyester (C) containing 2% by weight of N,N'-hexamethylenebisstearic acid amine 10

The intrinsic viscosity (η) of the polyesters (A) and (B) was 0.63, and that of the polyester (C) was 0.62.

Film-forming process:

The thus obtained polyesters (A), (B) and (C) were blended in the compositions shown in Table 1 and the thus obtained blends were dried, melt extruded at 285°C and then cooled and solidified, all in the known ways, to obtain the amorphous sheets. 15

These amorphous sheets were first stretched 36 times the original length in the machine direction at 85°C by using an IR heater to make the birefringence index (Δn) 0.090 and then stretched 4.0 times in the transverse direction at 120°C. The thus stretched sheets were then subjected to single-stage or double heat-setting at various temperatures and relaxed in the widthwise direction at various relaxation rates in the course of heat-setting to obtain the 100 μ m thick biaxially stretched films. These films are referred to as the films of Examples 1-2 and Comparative Examples 1-3, respectively. 20

EXAMPLE 3

0.1 part by weight of N,N'-hexamethylenebisbehenic acid amide was directly added and mixed in 100 parts by weight of a 97:3 blend of polyesters (A) and (B), and the mixture was worked into a film by following the same process as Example 1. The thickness of the obtained film was 100 μ m. 25

The properties of the films obtained in the Examples and Comparative Examples are shown in Table 1.

As seen from Table 1, the films of Comparative Examples 1 and 3 were high in coefficient of static friction. Also, blocking took place when these films were wound up after film formation and at the time of slitting, making it difficult to wind up the films. The film of Comparative Example 2 was increased in haze and poor in transparency. 30

TABLE 1

	Example 1	Example 2	Comparative Example 1	Comparative Example 2	Comparative Example 3	Example 3
Composition (parts)	A 94 3 3	87 3 10	97 3 0	22 3 75	94 3 3	97 3 0
Content of organic lubricant (wt%)	0.06	0.2	0	1.5	0.06	0.1*
Heat-setting temperature (°C)	240	240	240	240	220	240
Film haze	ΔH 0.2 H_0^{100} 0.6 H_0^{100} 0.8	0.2 0.8 1.0	0.2 0.6 0.8	0.6 0.9 1.5	0.2 0.6 0.8	0.2 0.7 0.9
Coefficient of static friction	μ_s 1.0	0.8	1.8	0.5	1.5	0.9
Film properties	\bar{n} 1.6063 ΔP 0.164	1.6063 0.168	1.6065 0.167	1.6062 0.165	1.6030 0.162	1.6064 0.169
Durability of metallized film	0	0	0	X	0	0
Synthetic evaluation	0	0	X	X	X	0

*Directly added.

CLAIMS

1. A biaxially stretched polyester film having excellent transparency and slipperiness, containing from 0.005 to 1% by weight of an organic lubricant and having the following properties after biaxial stretching and heat-setting:

$$\begin{aligned} \Delta H &\leq 0.3\% & (1) \\ H_0^{100} &\leq 0.9\% & (2) \\ \mu_s &\leq 1.2 & (3) \end{aligned}$$

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wherein Δ is the surface haze of the biaxially stretched film, H_0^{100} is the internal haze of the film as calculated based on the 100 μm film thickness, and μ_s is the coefficient of static friction of the film.

2. A biaxially stretched polyester film as claimed in claim 1, wherein the average refractive index (\bar{n}) of the film after biaxial stretching and heat-setting is in the range of from 1.640 to 1.670.

3. A biaxially stretched polyester film as claimed in claim 2, wherein the average refractive index (\bar{n}) of the film after biaxial stretching and heat-setting is in the range of from 1.650 to 1.670.

4. A biaxially stretched polyester film as claimed in any one of the preceding claims, wherein the content of the organic lubricant is in the range of from 0.05 to 0.5% by weight.

5. A biaxially stretched polyester film as claimed in any one of the preceding claims, wherein the surface orientation of the film after biaxial stretching and heat-setting is in the range of from 0.155 to 0.175.

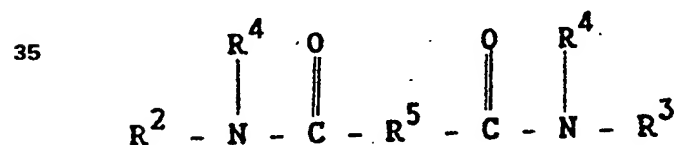
6. A biaxially stretched polyester film as claimed in any one of the preceding claims, wherein the thickness of the film is in the range of from 3 to 500 μm .

7. A biaxially stretched polyester film as claimed in any one of the preceding claims, wherein the organic lubricant is a compound of formula (4) or formula (5):

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- wherein R^1 is an alkylene group having from 1 to 12 carbon atoms, R^2 and R^3 are each independently an alkyl group having from 8 to 30 carbon atoms, R^4 is a hydrogen atom or a methyl group, and R^5 is an alkylene group having from 1 to 12 carbon atoms or a divalent hydrocarbon residue having an aromatic ring or aliphatic ring.

8. A biaxially stretched polyester film as claimed in claim 7, wherein the organic lubricant of formula (4) is bisamide compound obtained by reacting an alkylene-diamine such as ethylenediamine, propylenediamine, butylenediamine, hexamethylenediamine, octamethylenediamine, decamethylenediamine or dodecamethylenediamine with an aliphatic acid such as stearic acid, capric acid, decanic acid, lauric acid, myristic acid, palmitic acid, arachidinic acid, behenic acid, oleic acid, elaidic acid, montanic acid or carnaubic acid.

9. A biaxially stretched polyester film as claimed in claim 7, wherein the organic lubricant of the formula (5) is a bisamide compound obtained by reacting an alkyl-monoamine such as octylamine, decylamine, pentadecylamine, heptadecylamine, octadecylamine or dodecylamine with a carboxylic acid such as cyclohexanedicarboxylic acid, succinic acid, adipic acid, sebacic acid, terephthalic acid, isophthalic acid, phenylethyldicarboxylic acid, phenylethanedicarboxylic acid, naphthalenedicarboxylic acid or biphenyldicarboxylic acid.

10. A biaxially stretched polyester film as claimed in any one of the preceding claims, wherein the total haze (H_t^{100}) of the film as calculated based on the 100 μm film thickness is not more than 1.2%.

11. A biaxially stretched polyester film as claimed in any one of the preceding claims, wherein the coefficient of kinetic friction (μ_d) of the film is not greater than 1.2 and smaller than the coefficient of static friction (μ_s) of the film.

12. A recording material comprising a biaxially stretched polyester film as claimed in any one of claims 1 to 11, and at least one of a photosensitive layer, an emulsion layer, a transparent conductor layer having a photoconductor layer and a photosensitive layer laminated on the polyester film.

13. A transparent electrode comprising a biaxially stretched polyester film as claimed in any one of claims 1 to 11, and a transparent conductor layer laminated on said film.

14. A biaxially stretched polyester film as claimed in claim 1 and substantially as hereinbefore described with reference to the examples.

15. A recording material as claimed in claim 12 and substantially as hereinbefore described with reference to the examples.

16. A transparent electrode as claimed in claim 13 and substantially as hereinbefore described with reference to the examples.

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